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Effect of dissolved oxygen concentrations on the operational efficiency of the powdered activated carbon-membrane bioreactor (PAC-MBR) and single membrane bioreactor (MBR)

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ABSTRACT

Two experimental devices, namely, a powdered activated carbon (PAC)–membrane bioreactor (MBR) system and a single MBR system, were simultaneously operated under identical conditions. The changes in the dissolved oxygen (DO) concentration were observed in three phases. The removal efficiency of conventional pollutants in the two reactors was comparatively analysed; results indicated that the removal efficiency of the PAC-MBR system is greater than that of a single MBR system because the addition of PAC enhances the biodegradation of activated sludge, which is treated as a carrier for the adherence and growth of microorganisms and the adsorption of harmful substances. The DO concentration exerted lesser effects on the operational efficiency of PAC-MBR than on MBR. Moreover, the changes in the concentrations of proteins and polysaccharides in the two reactors were also examined. The protein concentration significantly changed, but that of polysaccharides did not change.

Keywords: Dissolved oxygen; MBR; PAC-MBR; Biodegradation; EPS

1.Introduction

Membrane bioreactors (MBRs) have been widely applied as a popular treatment method because of various advantages, including the enhanced enrichment and breeding of microorganisms, the improved removal efficiency of refractory organics and the good effluent water quality because of the direct solid–liquid separation by membrane filtration [1]. However, their widespread applications have been severely limited by membrane fouling. Shane Trussella [2] has reported that the increased sludge concentration leads to a decrease in membrane fouling, thus indicating that a high number of bacteria can be useful for the removal of pollutants but is not useful for permeability in MBRs. Kim [3] has reported different compositions of microbial community structures and extracellular polymeric substances (EPS) under high dissolved oxygen (DO) (>6.0 mg/L) and low DO (<0.1 mg/L) concentrations in MBRs. Microbial community structures and EPS are important operating parameters that can improve the control of membrane fouling. In addition to these parameters, other operational conditions (such as hydraulic retention time (HRT), sludge retention time (SRT), sludge concentrations and frequency of backwashing) have also attracted attentions [4,5]. By contrast, several additives have been added into MBRs to increase membrane permeability. The enhanced filterability in MBRs with the addition of fouling reducers is related to the increase in the floc size and the decrease in the concentration of soluble foulants (i.e., soluble microbial products (SMP) or EPS) in the bulk phase [6]. Yoon [7] used modified cationic polymers (e.g., Permacare MPE50TM and MPE51 TM) to reduce biopolymer levels and increase the particle size for

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controlling membrane flux. Koseoglu [6] reported improved performance with the addition of cationic polymer KD452 compared to seven other chemical additives in terms of the extent of SMP removal and the required dosage. Khan [8] compared the filtration performance and sludge characteristics of conventional submerged MBR versus hybrid MBRs with the addition of cationic polymer and powdered activated carbon (PAC) and revealed that the addition of PAC to MBR is the most suitable option related to the combination of flocculation and adsorption. Hence, PAC demonstrates promise for change membrane fouling.

PAC is a carbonaceous material with a developed pore structure and a large specific surface area of 1000–1500 m^2/g. Its density is less than that of water, and it exhibits excellent adsorption performance, particularly for the dissolved organic matter (DOM) in wastewater. Furthermore, it is a cost-effective material that can be easily obtained. Thus, several studies have reported the addition of PAC in the MBR. Hu [9] compared the effect of different PAC dosages on the removal efficiencies of organic matter and ammonia, as well as the control of membrane fouling, in a pilot-scale PAC-MBR treating micro-polluted surface water and found that an initial PAC dosage of 2 g/L is optimal, with renewal after 21 d of continuous operation. Shao [10] reported the application of intermittent aeration and low SRTs to the PAC-MBR for the treatment of micro-polluted surface water because they can provide sufficient oxygen for bacteria and because low SRTs can be treated as an effective factor for decreasing membrane fouling. Furthermore, Hu [11] found that the addition of PAC into an activated sludge system improves the biomass and sludge settleability and changes the microbial community structures.

In this study, PAC was added into an MBR to examine its effect on activated sludge under different DO conditions. Two goals were investigated: (i) whether the addition of PAC could decrease membrane fouling be lower than none of this in reactor, and (ii) whether the enhanced removal efficiency of pollutants was related to the improved biomass.

2. Materials and methods

2.1. Experimental set-up

Two identical lab-scale MBRs were built (Fig. 1). Each MBR with a total working volume of 0.5 L was operated in parallel and fed with artificial simulated wastewater for 180 d. PAC was added into one reactor at a fixed concentration of 2 g/L while simultaneously varying other concentrations. The overall HRT of the MBRs was 4 h, and the SRT was 180 d. The MBRs were equipped with two immersion-type hollow-fibre PVDF fasciculation membranes with a nominal pore size of 0.01 µm and a surface area of 0.1 m^2 each. The membrane flux was 1.25 $L/m^2\cdot h$, and the flux of backwash water was 4.5 $L/m^2\cdot h$. Coarse bubble aerators were placed below the membranes to provide 24 h continuous aeration, which was required to maintain the DO concentrations for satisfying the requirements of microorganisms and enabling the mixed liquid to maintain turbulent conditions for decreasing membrane fouling in the system. The change tendency of DO concentrations was divided into three phases, which were 2.41–1.82 (Phase I), 1.82–0.86 (Phase II) and 0.86–0.32 mg/L (Phase III), respectively. The filtration

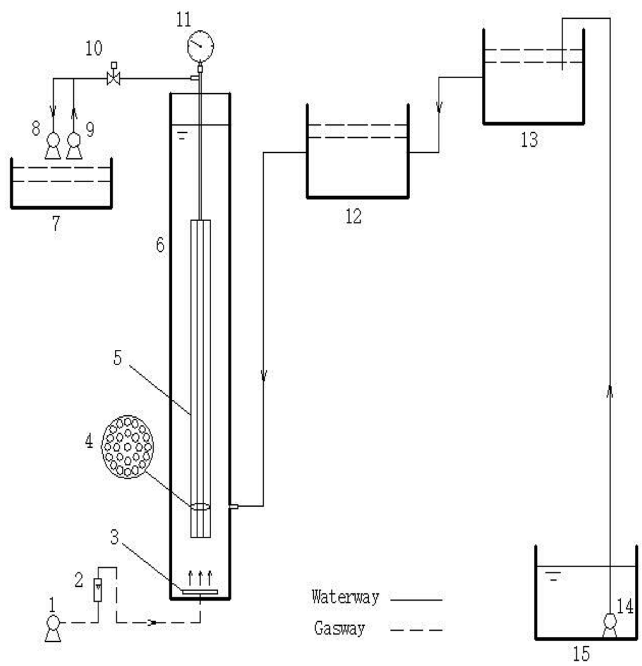

Fig. 1. The experimental facility (1 – Air pump; 2 – Gas flow meter; 3 – Air diffuser; 4 – Cross section; 5 – Membrane; 6 – Bioreactor; 7 – Sink tank; 8 – Suction pump; 9 – Backwash pump; 10 – Relays; 11 – Vacuum gauge; 12 – Constant level tank; 13 – High water tank; 14 – Submersible pump; 15 – Water distribution tank).

and backwashing time set by the PLC system and time were approximately 10 and 1 min, respectively. The reactor was operated at 22°C ± 3°C with temperature control via an electric heating rod.

2.2. Artificial simulated wastewater

The artificial simulated wastewater was considered experimental water. $C_6H_{12}O_6$ and CH_3COOH were used as comprehensive carbon sources, NH_4Cl was used as the nitrogen source and K_2HPO_3 and KH_2PO_3 were used as the phosphorus sources. Furthermore, appropriate amounts of $MgSO_4\cdot 7H_2O$, $CaCl_2$, $CuSO_4\cdot 5H_2O$, $MnCl_2\cdot 2H_2O$, $ZnSO_4\cdot 7H_2O$ and essential microelements for microbial growth were added. $NaHCO_3$ was used as the pH regulator. pH was maintained at 7–7.5. Chemical oxygen demand (COD), ammoniacal nitrogen (NH_4^+-N) and total phosphorus (TP) were simultaneously maintained at approximately 450, 12 and 2.0 mg/L, respectively.

2.3. Inoculated sludge

The inoculated sludge in this study was obtained from the sludge-thickening tank at Wenchang Urban Sewage Treatment Plant in Heilongjiang, China. The dark brown sludge was slightly acidic at a pH of 6.2. Approximately, 1 L of the sludge was placed in a beaker, and nutrients were added. The recovered sludge was inoculated after 24 h of aeration. The volume of the inoculated sludge was 1/3 of the working volume of the reactor. Numerous microorganisms, which exhibited high activity in the inoculated sludge, were observed by microscopic examination. The microorganisms exhibited a relatively clear (transparent) appearance and structure. The VSS/SS was 0.58.

2.4. Analytical methods

COD, ammonia nitrogen (NH_3-N), TP, total nitrogen, MLSS and MLVSS were measured according to methods reported previously [12–14]. Protein was analysed [15] by Coomassie blue staining, and polysaccharide was analysed by the anthrone sulphuric acid spectrophotometric method [16]. The 3D excitation–emission matrix (EEM) was analysed using a fluorescence spectrophotometer (JASCO, FP-6500).

2.5. Characteristics of the powdered activated carbon

The specific surface area, pore size and pore volume of PAC were measured by nitrogen adsorption at −196°C (liquid nitrogen temperature) by using a specific surface area analyser (ASAP2020, Micromeritics, US), and the average particle volume was measured using a laser particle size analyser (Mastersizer 2000, Malvern Instruments, UK). Table 1 summarizes the characteristics of PAC.

3. Results and discussion

3.1. Decontamination efficiency of two MBRs

3.1.1. Removal efficiency of organic matter in two MBRs

The average COD removal efficiency for the PAC-MBR system was slightly greater than that of the single MBR system (Fig. 2a). Hence, the addition of PAC is thought to improve the MBR removal efficiency of organic matter. This result is in agreement with those reported previously [17–20]. Han [21] has reported that the COD removal efficiency for the PAC-MBR system increases with the addition of 0.1–2 g/L of PAC. In the PAC-MBR system, PAC served as the carrier, and activated sludge adhered to its surface; thus, some biological activated carbon (BAC) flocs are formed, which exhibit excellent biological activity. Furthermore, the performance of sludge is enhanced, and biodegradability is improved. Given the constant accumulation and compression of BAC on the membrane module surface with the increase in operation time, dense filter cake and

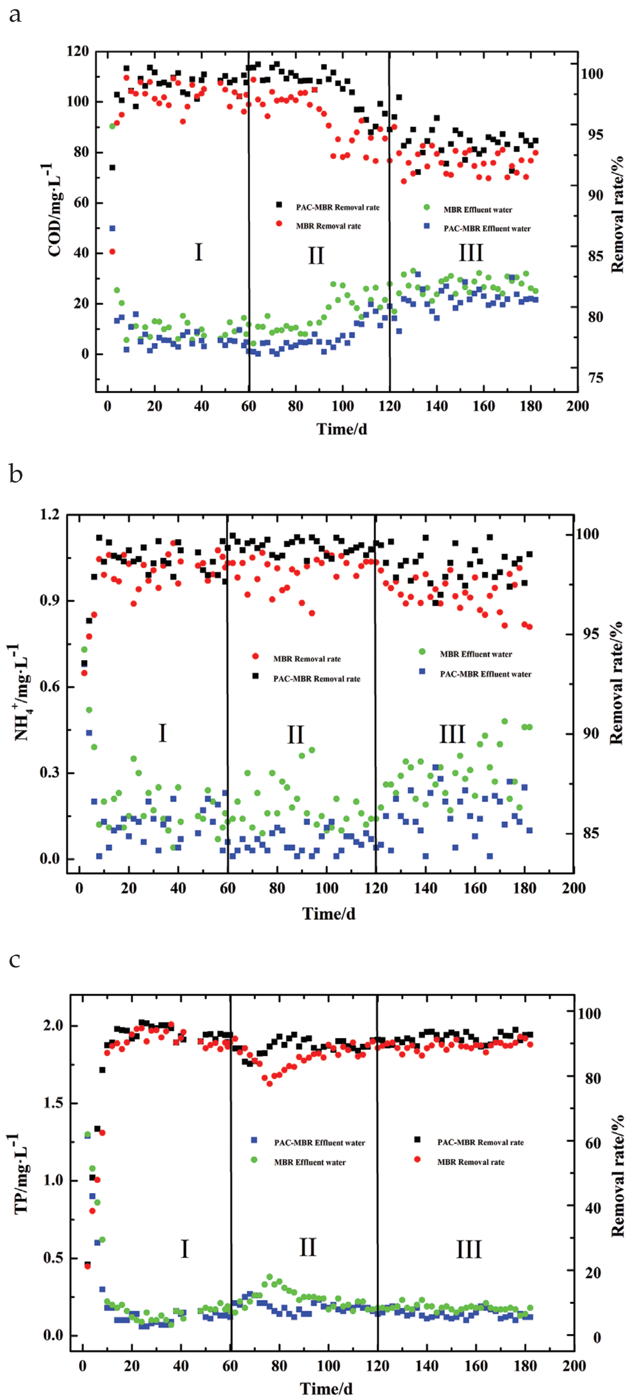

Fig. 2. The operational efficiency of two MBRs: (a) The COD removal efficiency; (b) The NH_4^+-N removal efficiency; (c) The TP removal efficiency.

gel layers were formed, which were well intercepted and degraded to soluble organic molecules in water; moreover, the ultrafiltration membrane also exhibited good interception ability. The addition of PAC could also adsorb soluble organic molecules in water. Hence, the water quality of the effluent improves [22].

Table 1
The characteristics of PAC

Parameter	Unit	PAC
Raw material		Wood/peat
Average particle volume	μm^3	62.9117
Surface	m^{2}*g^{-1}	619.6968
Pore volume	cm^{3}*g^{-1}	0.246713
Pore size	nm	4.9601

With the gradual decrease in the DO concentration, the removal efficiency of organic matter decreased in the two MBRs. The reactors were under limiting oxygen conditions, and the DO concentration (aeration) decreased moving from Phase I to III in the system for the purpose of decreasing the oxygen transfer rate. The aerobic bacterial activity was inhibited, and the microbial metabolic rate decreased. Thus, the degradation rate of the organic carbon source decreases. However, the activity of the facultative anaerobic bacteria and anaerobic bacteria progressively increased, and these bacteria exhibited specific removal efficiency for the organic matter in wastewater. Hence, the removal rate is maintained at greater than 90%. When the operation of the two MBRs was stable at different stages, the removal rates for the PAC-MBR system were 98.48%, 97.06% and 93.31%, whereas those for the MBR system were 97.27%, 93.76% and 91.76%. Therefore, the COD removal efficiency of PAC-MBR is better than that of the single MBR during different DO concentrations because the addition of PAC leads to the enhancement in the organic matter removal efficiency of a bioreactor. Moreover, the decreasing trend of the PAC-MBR system was less than that of the single MBR system because the adsorption of PAC led to the generation of a large number of oxygen molecules on the surface, which was beneficial for microbial metabolism.

3.1.2. Removal efficiency of ammonia nitrogen in the two MBRs

The average NH_3-N removal efficiency of PAC-MBR was slightly greater than that of the single MBR (Fig. 2b). Thus, the removal efficiency of the PAC-MBR system is slightly higher. A large number of microorganisms were enriched because of the efficient interception of MBRs, which was beneficial in strengthening the proliferation of nitrifying bacteria; in addition, it was more conducive to the survival and reproduction of microorganisms in the presence of a sufficient DO concentration and an appropriate pH; therefore, MBRs exhibit excellent removal efficiency for NH_3-N. With the addition of PAC into the reactor, good growth sites were obtained for microbes such that the number of nitrifying bacteria in the entire system was greater than that in the MBR, thereby improving the NH_3-N removal efficiency [23,24]. Furthermore, PAC can adsorb some toxic substances, which could affect the normal operation of nitrification; hence, the biological activity and stability of the system are improved [25,26]. However, several studies have reported that the addition of PAC may not affect the removal of nitrogen [27–29].

In the water, NH_3-N existed as free ammonia and ammonium ions. Small inorganic molecules can freely pass through membrane pores. Thus, the removal efficiency of NH_3-N mainly depends on biodegradation rather than the membrane interception in the MBR system. The NH_3-N removal efficiency was not significantly affected with the change in the DO concentration during the entire process of the reactor operation (Fig. 2b). The aerobic and facultative denitrifying bacteria simultaneously existed in the reactor, and their antagonistic relationship exhibited a dynamic balance to maintain a removal rate of greater than 95%. Moreover, when the operation of the two MBRs was stable at different stages, the removal rates of the PAC-MBR system were 98.72%, 99.34% and 98.47%, whereas those of the MBR system were 98.44%, 98.33% and 96.82%. Therefore, the NH_3-N removal efficiency of PAC-MBR is better than that of the single MBR during different DO concentrations because PAC provides good sites for the growth of nitrification bacteria [23,24]. The amount of bacteria in the PAC-MBR system is greater than that of the single MBR system under the same operational conditions.

3.1.3. Removal efficiency of phosphate in the two MBRs

Biological phosphorus removal occurs when the phosphorus-accumulating organisms (PAOs) combine the anaerobic phosphorus release with the aerobic phosphorus to complete the treatment. Phosphorus is absorbed to a greater extent under aerobic conditions compared with that released under anaerobic conditions. Hence, the phosphorus removal in wastewater discharge sludge comprises a large number of poly-β-hydroxybutyrate. However, the SRT in this experiment was infinite. Thus, the phosphorus removal only depends on the microbial assimilation. The average TP removal efficiency of PAC-MBR was slightly greater than that of the single MBR. Continuous 24 h aeration is thought to be advantageous for the phosphate uptake of poly-P bacteria under aerobic conditions. However, at the same time, the anaerobic environment was limited in the reactor, and no sludge was discharged during the entire operation; thus, the two MBRs exhibit low TP removal efficiencies, which mainly relies on the assimilation of microorganisms. With the continuous operation of the reactor, the removal efficiency started to stabilise possibly because the operational process leads to the increase in the sludge concentration and microbial biomass, thus leading to the combination of biodegradation and adsorption; in addition, the typical metabolism of microorganisms requires the use of phosphorus to synthesise its own material; thus, the TP removal efficiency of MBRs achieves a dynamic balance [30–32]. With the addition of PAC into the reactor, a number of growth sites for microorganisms were available; hence, microbial enrichment occurs. A high phosphorus concentration was required for microbes in the PAC-MBR system.

The TP removal rate rapidly increased because of the rapid microbial proliferation in the presence of high DO concentrations via the adsorption of phosphate in the reactor to synthesise their own desired substance. Microbial growth gradually decreased with the reactor operation, and their death and growth afforded a dynamic balance. Thus, the removal rate reaches a stable state. The phosphorus removal in Phase II decreased because of the decrease in the DO concentration. The activity of aerobic PAOs is inhibited. However, with the reactor operation, aerobic PAOs adapted to the surrounding environment, and the activity gradually recovered. The phosphate removal was maintained at greater than 85%. Moreover, when the operation of the two MBRs was stable at different stages, the removal rates of the PAC-MBR system were 92.68%, 89.51% and 91.88%, whereas those for the MBR system were 91.97%, 87.65% and 89.73%. The phosphate removal efficiency of PAC-MBR is better than that of the single MBR during different DO concentrations.

3.2. Change in the sludge concentration in the two MBRs

With the decrease in the DO concentration in the two reactors, the sludge concentration increased (Fig. 3). First, a high amount of sludge was generated in the reactors because of the high interception of the ultrafiltration membrane, which was possibly responsible for the increased sludge concentration. Second, the SRT was infinite; that is, the sludge was not discharged unless necessary experimental dates needed testing to have a considerable amount of sludge in the two MBRs. Hence, the sludge concentration inevitably increases. Third, the metabolism and endogenous respiration of microorganisms decreased with the DO concentration. Furthermore, the concentration of sludge in PAC-MBR was greater than that in the single MBR. In the PAC-MBR system, the added PAC can be treated as a carrier for the adherence and growth of microorganisms, as well as for the adsorption of harmful substances. PAC-MBR provides a better growth environment for microorganisms; hence, the amount of sludge is greater than that of the single MBR.

3.3. Variation in the EPS content and components

3.3.1. EPS content

The EPS consists of a number of high-molecular-weight substances and was considered the main factor affecting the membrane fouling of MBR [33–35]. In particular, the protein content was greater than that of polysaccharide. Fig. 4 shows the change in the EPS content under different DO concentrations. The protein concentration decreased with the DO concentration in the single MBR system because a considerable amount of proteins, which are classified as extracellular enzymes, were released because of the high microbial activity and metabolism under high DO (i.e., high aeration) conditions. Hence, the protein concentrations at different stages follow the order of Phase I > Phase II > Phase III. However, the concentrations of proteins in the PAC-MBR system at different stages followed the order of Phase I > Phase III > Phase II. The activity of facultative aerobic bacteria and anaerobic bacteria gradually increased because the addition of PAC was beneficial to the reproduction of microorganisms even if the activity of the aerobic bacteria decreased with decreasing DO concentrations. Thus, a high protein concentration is observed in Phase III. Moreover, hydraulic shear stress was enhanced, and the sludge flocs were destroyed under high DO concentrations (i.e., high aeration rate); hence, a large amount of EPS is released. The concentration of proteins in the single MBR was greater than that in the PAC-MBR system at each stage because the addition of PAC combined with the sludge flocs exhibit a buffering efficiency on the hydraulic shearing force. Polysaccharide can be consumed as a carbon source by microorganisms for metabolism. The single MBR system exhibited a low variation in the polysaccharide content, and the consumption and formation of polysaccharides exhibited a dynamic balance. The change trend in polysaccharides was similar to that of proteins in the PAC-MBR system. Moreover, the PN/PS ratio decreased with decreasing DO concentration because of the small change in the polysaccharide content and the continuous decrease in the protein content.

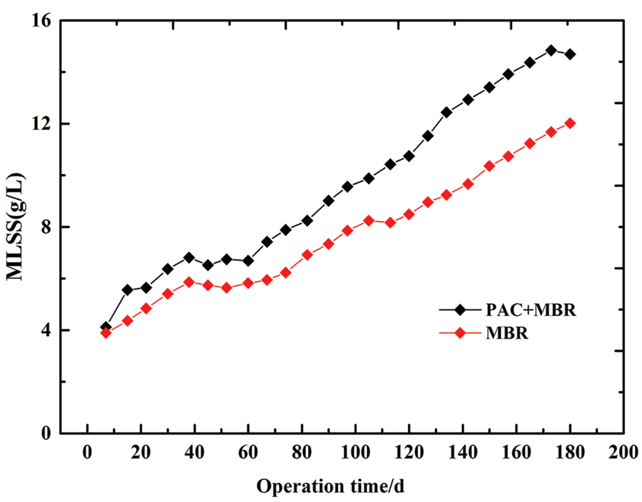

Fig. 3. The change of sludge concentration in two reactors.

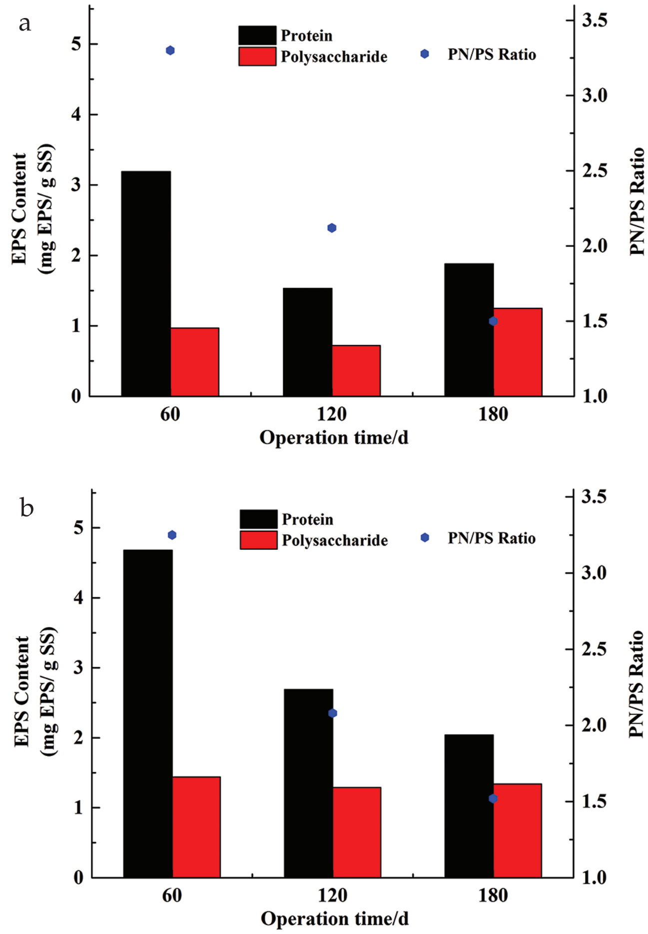

Fig. 4. The EPS content in different reactors (a) The EPS content of activated sludge in PAC-MBR; (b) The EPS content of activated sludge in MBR.

3.3.2. EPS components

Three-dimensional EEM fluorescence spectroscopy has been widely employed as a high-sensitivity method. A small number of samples were analysed by a rapid, simple analysis method, and each component of the DOM was effectively distinguished on the basis of its fluorescence properties [36–39]. Thus, spectroscopy is employed to understand the specific fluorescence characteristics of protein-like substances. Furthermore, spectroscopy was employed to completely classify and measure the concentration of phytoplanktons on the surface body for

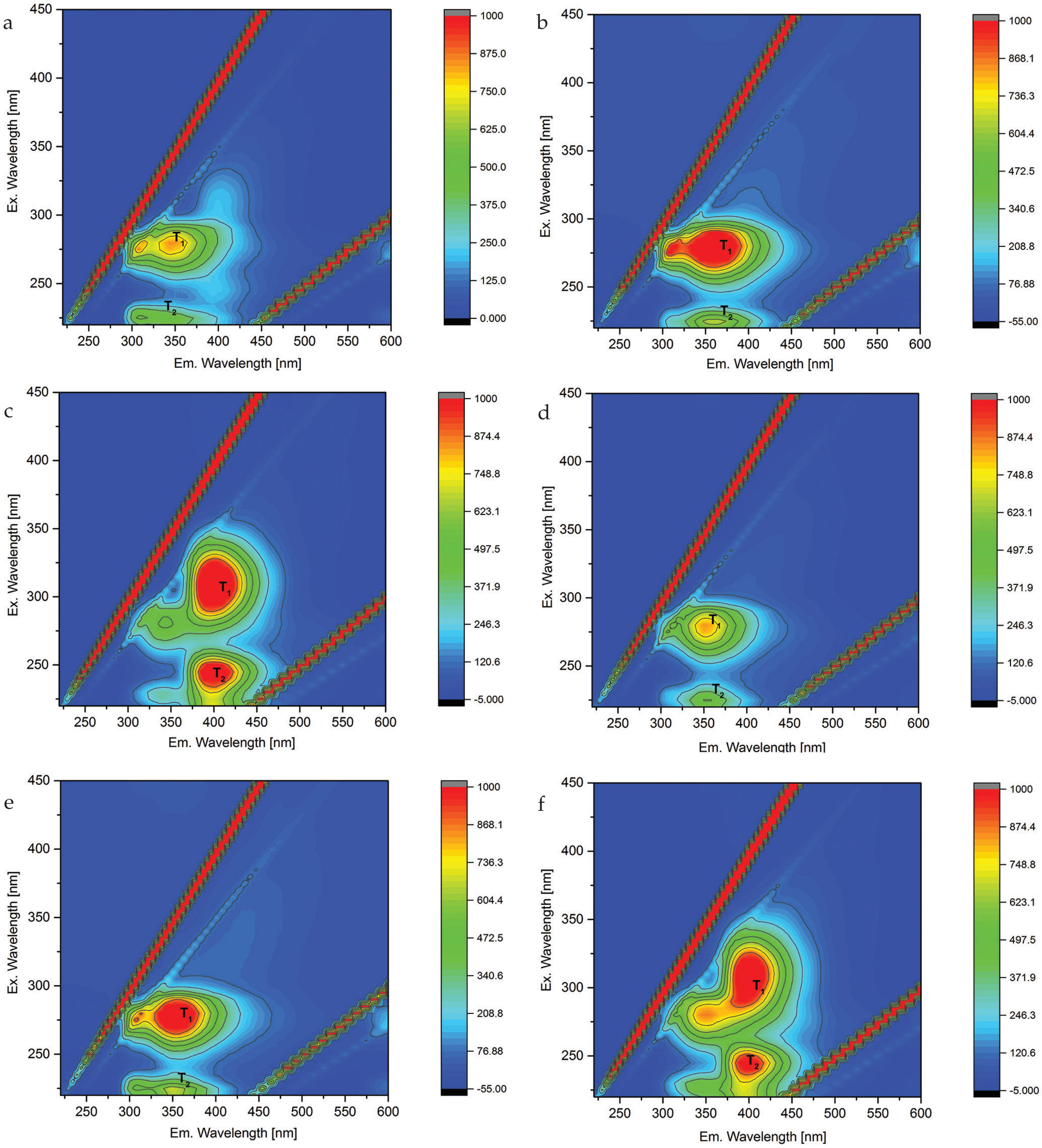

Fig. 5. 3D-EEM spectra of EPS in activated sludge of two reactors: (a) 60 d in MBR, (b) 120 d in MBR, (c) 180 d in MBR, (d) 60 d in PAC-MBR, (e) 120 d in PAC-MBR, (f) 180 d in PAC-MBR.

the characterisation of the DOM in surface waters and to analyse the characteristics of organic membrane foulants in a submerged MBR with pre-ozonation. The peak intensities of T1 and T2 (protein-like substances) were significantly enhanced with the decrease in the DO concentration in the single MBR system (a→c) and PAC-MBR system (d→f). The total amount of protein increases. However, the sludge concentration also increased (Fig. 5). Therefore, the yield of protein in the unit mass sludge decreased and exhibited a negative correlation with the peak intensity of T.

4. Conclusion

In this study, two MBR systems were first compared, and the addition of PAC was found to enhance the operational efficiency of MBR because BAC flocs were formed in the PAC-MBR system via the addition of PAC as the carrier; hence, the biodegradation of sludge is improved. Second, the removal efficiency of organic matter, NH_3-N and TP were affected by the decrease in the DO concentration. The overall operation of the two reactors did not significantly change. Thus, the stability of the MBR system mitigates the efficiencies, which is related to the change in the external environment. Third, the protein concentration significantly changed with decreasing DO concentrations because the DO concentration was proportional to the hydraulic shear force, and the sludge floc structure endured the stress. Finally, the PAC-MBR system mitigated membrane fouling and exhibited higher biodegradation ability than that exhibited by the single MBR system. Moreover, the addition of PAC led to the adsorption of harmful substances that affected the growth of activated sludge. Hence, PAC-MBR is suitable for practical engineering applications.

Acknowledgments

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